

# PATENT SPECIFICATION

692,586



Date of Application and filing Complete Specification: Nov. 16, 1950.

No. 28087/50.

Application made in Switzerland on Nov. 16, 1949.

Complete Specification Published: June 10, 1953.

**Index at acceptance:**—Class 2(v), RP8(a: c4b), RP8c8(b: c), RP8c(9: 12b), RP8c13(a: c), RP8d1(a: b), RP8d2(a: b2), RP8d(4: 5), RP8k(8: 9), RP8p1(c: e5: f), RP8p2(a1: b), RP8p6(c: d: f), RP8s1, RP9(a: c4b), RP9c8(b: c), RP9c(9: 12b), RP9c13(a: c), RP9d1(a1: b1: x), RP9k8, RP9p1(c: e5: f), RP9p2(a1: b), RP9p6(c: d: f), RP9s1.

## COMPLETE SPECIFICATION

### Process and Material for Taking Dental and Other Impressions

We, FARBENFABRIKEN BAYER, a German Company recognised under German Law, of 22c, Leverkusen-Bayerwerk, Germany, do hereby declare the invention, for which we pray that a patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following statement:—

10 The present invention relates to a new process and a new material for taking impressions, especially for dental purposes.

15 The new process comprises the use of polymerizable vinyl compounds which form gels in their polymerized state with solvents. Impressions are taken according to the present invention by means of pasty mixtures of polymerizable vinyl compounds with appropriate solvents. A polymerization catalyst is added to the mixture, when about to be used, to initiate polymerization and the mixture is then contacted with the object of which

20 an impression is to be taken until setting by further or substantially complete polymerization with gel formation has occurred.

25 The pasty condition required may be achieved in various ways. The simplest is to make the solution of the vinyl compound in the solvent into a paste by means of solid fillers. However, there are other ways, such as to add to the solution substances increasing its viscosity, or to cause or allow the polymerization catalyst to act, before applying the mixture, to such a degree that the desired pasty condition is attained by the progress of

30 35 40 the polymerization. Any of these means may be used in combination.

Especially suitable solvents for dental

impression materials are water or strongly hydrophilic organic solvents, such as glycerine or aqueous solutions of glycerine. Suitable polymerizable vinyl compounds forming gels in their polymerized state in water, glycerine and the like are acrylic acid, methacrylic acid and mixtures thereof with fumaric acid and maleic acid. Instead of these acids, their salts, amides, hydroxyalkylesters or other water-soluble derivatives may be used.

The polymerization catalysts are preferably such as will assure completion of the polymerization within a short time. The activation and reaction conditions for dental purposes should be such that the polymerization temperature does not exceed 45° C. Such catalysts are available, for instance, in the so-called "Redox-systems," i.e. mixtures of oxidizing and reducing agents. The reaction velocity may be promoted by the application of ultraviolet rays.

Suitable mineral fillers are various inorganic pigments, calcium sulphate ( $\text{CaSO}_4$ ,  $\text{CaSO}_4 \cdot 1/2\text{H}_2\text{O}$ ,  $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$ ) titanium dioxide, aluminium oxide, bentonite and talcum.

Substances used for increasing the viscosity of the solution may be silica gels, especially those capable of swelling in water. As examples of other such substances there may be mentioned carbohydrates, such as agar-agar and dextrine; cellulose derivatives soluble in the solvent applied, for instance methyl cellulose; salts of alginic acid or polyacrylic acid; and polymerates of the polymerizable vinyl compounds used according to the invention.

The impression masses may contain the

customary additions such, for example, as dyestuffs, disinfecting agents, flavouring materials and fibres.

In carrying out the invention it is preferred to use polyfunctional products as polymerizable vinyl compounds, that is to say organic compounds containing more than one vinyl group. These products may be used alone or in combination with monofunctional vinyl compounds. Suitable polyfunctional vinyl compounds are, for instance, polyamides of acrylic acid or methacrylic acid, and organic compounds containing at least two amino groups, such as ethylene diamine or hexamethylene diamine. Another example is "triacrylformal," i.e. the reaction product of acrylonitrile and anhydrous formaldehyde (cf. Chemische Berichte vol. 81 (1948) p. 527). These polyfunctional vinyl compounds promote gel formation and improve the elastic properties of the polymerized material. They are preferably employed in admixture with monofunctional vinyl compounds.

It will be understood that the invention may be performed in various ways and that many modifications and changes may be made owing to the variety of vinyl compounds and fillers which may be used. There is therefore ample scope for varying combinations adapted to meet any practical requirement.

The following examples, in which the parts mentioned are by weight, are given for the purpose of illustrating the invention.

#### EXAMPLE 1.

5 parts of a 16.5 per cent. aqueous solution of monomeric acrylamide are thoroughly stirred with 3 parts of a 10 per cent. aqueous solution of polyacrylic acid, 2 parts of gypsum, 0.5 part of a 10 per cent. aqueous solution of ethylene diacrylamide and 1.5 parts of magnesium oxide. By adding to the past thus obtained 0.1 part of potassium persulphate and 0.2 part of triethanolamine in some water with vigorous stirring polymerization is soon initiated with a slight increase of temperature. The compounded mass thus obtained may be placed on an impression tray for application to the mouth before polymerization is complete and before the material has set. Gelation and setting of the impression takes place within a few minutes.

#### EXAMPLE 2.

4 parts of calcium carbonate and 3 parts of magnesium carbonate are made into a stable paste with 10 parts of a 16.5 per cent. aqueous acrylamide solution, 6 parts of a 10 per cent. aqueous polyacrylic acid solution, 1 part of a 10 per cent. aqueous solution of ethylene diacrylamide and 0.8

part of a 10 per cent. aqueous solution of triethanolamine. An addition of 0.2 part of potassium persulphate initiates polymerization.

#### EXAMPLE 3.

70 10 parts of talcum, 10 parts of a 16.5 per cent. acrylamide aqueous solution, 0.3 part of magnesium carbonate, 0.1 part of triacrylformal, 2 parts of a 10 per cent. polyacrylic acid aqueous solution, are 75 made into a paste with stirring with gradual or intermittent addition of 4 parts of water. For initiating polymerization 0.5 part of potassium persulphate and 0.3 part of formamidine sulphonic acid 80 are added.

#### EXAMPLE 4.

10 parts of talcum, 10 parts of a 16.5 per cent. aqueous acrylamide solution, 0.13 part of magnesia, 0.2 part of triacrylformal, 0.5 part of gelatine and 2 parts of a 10 per cent. polyacrylic acid aqueous solution are made into a stable paste with the addition of 4 parts of water. Gelation of the paste by means of 90 polymerization is initiated by adding 0.3 part of potassium persulphate and 0.2 part of formamidine sulphonic acid.

#### EXAMPLE 5.

95 15 parts of talcum and 2 parts of silica gel are ground together with 0.05 part of triacrylformal, 4 parts of acrylamide and 25 parts of a 1.5 per cent. agar aqueous solution. 0.1 part of triethanolamine and 0.2 part of potassium persulphate are 100 added to initiate polymerization.

#### EXAMPLE 6.

105 20 parts of talcum are made into a stable paste with 20 parts of 75 per cent. glycerine, 4 parts of acrylamide, 0.5 part of triacrylformal and 0.15 part of triethanolamine. Gelation of the paste is initiated by adding 0.5 part of potassium persulphate.

#### EXAMPLE 7.

110 A mixture of 20 parts of gypsum, 20 parts of talcum, 5 parts of acrylamide and 0.7 part of formamidine sulphonic acid is stirred with 25 parts of water. The mixture sets within 5 minutes when 0.7 part of potassium persulphate is added. It may be mentioned, by way of comparison, that a mixture of gypsum, talcum and water alone would set to a brittle mass.

#### EXAMPLE 8.

120 20 parts of talcum, 5 parts of 69 per cent aqueous solution of acrylic acid, 0.07 part of triacrylformal, 3.5 parts of silica gel and 0.5 part of formamidine sulphonic acid are made into a paste with 20 parts of water with stirring. After adding 0.5 part of potassium persulphate the paste polymerizes on the impression mould within a few minutes to form an 130

elastic accurately shaped impression.

**EXAMPLE 9.**

A mixture of 3.8 parts of acrylamide, 8 parts of zinc oxide, 6 parts of talcum, 15 parts of highly active finely divided silica gel, 2.5 parts of sugar, 0.7 part of calcium oxide, 0.15 part of triacrylformal, 0.2 part of filaments of high-polymeric, linear polyamides or other linear high polymers (thin individual filaments cut to about 0.2 cm. length or twisted cotton threads of the same length) and 0.5 part of triethanolamine hydrochloride is stirred with 55 cc. of water of 18—22° C. until a homogeneous paste has been formed. Thereupon 0.25 part of potassium persulphate is added to the paste which is thoroughly mixed and kneaded with a spatula, placed on the impression tray and applied to the mouth within  $\frac{1}{2}$  to 1½ minutes after the addition of the persulphate. The compounded material sets with slight heating (to 38° C.). The impression is taken from the mouth after 2—3 minutes by means of a slight pressure and subsequently cast with gypsum.

**EXAMPLE 10.**

5 parts of acrylamide and 0.15 part of triacrylformal are mixed with 20 parts of silica gel, 0.5 part of zinc oxide, 3 parts of sugar and 1.2 parts of triethanolamine. On kneading the mixture with 50 parts of water a paste is obtained which polymerizes with 0.25 part of potassium persulphate within a few minutes to an elastic gel.

**EXAMPLE 11.**

4 parts of acrylamide and 0.3 part of triacrylformal are stirred with 16 parts of silica gel, 8 parts of zinc oxide, 2 parts of triethanolamine, 20 parts of sugar, 15 parts of glycerine and 15 parts of water to a stable paste. Gelation of the paste is initiated by adding 1 part of potassium persulphate. The paste is applied to the object of which an impression is to be taken before a gel has formed, so that gel-formation takes place in contact with said object. The impression material is easily removed from the object when gel-formation is complete.

What we claim is:—

1. Process for taking an impression of an object for dental or other purposes, which comprises producing a pasty mix-

ture comprising a polymerizable vinyl compound, a solvent capable of forming a gel with the vinyl compound in its polymerized state, and a filler and/or a substance for increasing the viscosity of the solvent, adding a polymerization catalyst so as to initiate polymerization of the vinyl compound and contacting the pasty mixture with the object before gel-formation has taken place, said pasty mixture being set by further polymerization with gel-formation while in contact with the object.

2. Process as claimed in claim 1 in which the polymerizable vinyl compound is methacrylic acid, acrylic acid, or a salt or amide or hydroxyalkyl ester or other water-soluble derivative thereof, or a mixture of any of the foregoing.

3. Process as claimed in claim 2 in which the polymerizable vinyl compound contains fumaric acid, maleic acid, or an amide, hydroxyalkyl ester, a salt or another water-soluble derivative thereof.

4. Process as claimed in claim 1, 2 or 3 in which a monofunctional vinyl compound is used in admixture with a polyfunctional vinyl compound.

5. Process as claimed in claim 4, in which the polyfunctional vinyl compound is a polyamide of acrylic acid or methacrylic acid and an organic compound containing at least two amino groups, or is triacrylformal.

6. Process as claimed in any of claims 1—5, wherein a redox system is used as polymerization catalyst.

7. Process as claimed in any of claims 1—5 in which potassium persulphate is used as polymerization catalyst.

8. Process as claimed in any of claims 1—7 in which the solvent is hydrophilic such as water, glycerine, or a mixture thereof.

9. Material for taking dental or other impressions whenever prepared by a process as claimed in any preceding claim.

10. The process for taking impressions of objects, and the material therefor, substantially as described in any of the Examples.

**CARPMAELS & RANSFORD,**

Agents for Applicants,  
24, Southampton Buildings,  
Chancery Lane, London, W.C.2.